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<b>(21) International Application Number:</b> PCT/GB93/01473 <b>(22) International Filing Date:</b> 14 July 1993 (14.07.93)  <b>(30) Priority data:</b> 9216776.6                      7 August 1992 (07.08.92)                      GB  <b>(71) Applicant (for all designated States except US):</b> IMPERIAL CHEMICAL INDUSTRIES PLC [GB/GB]; Imperial Chemical House, Millbank, London SW1P 3JF (GB).  <b>(72) Inventor; and</b> <b>(75) Inventor/Applicant (for US only) :</b> TURRY, Bernard [GB/ GB]; 8 Spring Vale, Prestwich, Manchester M25 8SE (GB).  <b>(74) Agents:</b> DRAGGETT, Peter, Thornton et al.; ICI Group Patents Services Dept, P.O. Box 6, Shire Park, Bessemer Road, Welwyn Garden City, Hertfordshire AL7 1HD (GB).		<b>(81) Designated States:</b> AU, BR, CA, JP, KR, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> COMPOSITION AND USE  <b>(57) Abstract</b>  A composition comprising a hydroxyoxime together with the reaction product of an alkyl substituted cyclic anhydride and a hydrophilic compound containing hydroxyl and/or amino groups, a process which comprises treating at least part of the surface of a metal with the composition, a metal coated with the composition, and a lubricant composition comprising the composition and an oil or a grease. The reaction product of the alkyl cyclic anhydride may be the reaction product of polyisobutenyl succinic anhydride and ethanolamine. The hydroxyoxime may be 2-hydroxy-5-nonylbenzaldoxime or 2-hydroxy-5-nonylacetophenoneoxime. The composition may be used to inhibit corrosion of metals, particularly iron.		

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## COMPOSITION AND USE

This invention relates to a new composition, and the use of such a composition to inhibit the corrosion of metals, especially when incorporated into oils and greases, and thus to a process which comprises treating least part of the surface of a metal with the composition, a metal coated with the composition, and a lubricant composition comprising the composition and an oil or a grease.

In our published European Patent Application No 247728 there is disclosed a composition comprising an optionally substituted hydroxyoxime and an anhydride containing a long alkyl chain. The composition may be incorporated into an oil for the protection of metal surfaces against corrosion, and under most conditions, the composition provides extremely good protection of the metal surface against corrosion. However, when moisture is present the efficacy of the composition can be severely impaired, and the anti-corrosive properties imparted to the metal surface can be unacceptable.

The composition disclosed in EP 247728 can also be incorporated in to greases for the lubrication of metal surfaces which are in sliding or frictional contact. In most instances such grease formulations provide excellent antiwear characteristics on those metal surfaces. However, in the presence of moisture, the grease has been found to destructure under certain circumstances and the lubricating characteristic of the grease is lost thus resulting in greater wear and/or corrosion of the metal surfaces.

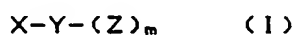
We have now found that improved corrosion inhibition and greater resistance to moisture can be obtained using a mixture of a hydroxyoxime compound together with the reaction product of a substituted cyclic anhydride and an organic moiety containing amino and/or hydroxyl groups.

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That such compositions have been found effective is surprising, since in most cases the individual components of the composition have been found to exhibit little, if any, anti-corrosion properties, when incorporated into to an oil and applied to a metal surface.

According to the present invention, there is provided a composition comprising

- a) a surface-active anhydride derivative of the general formula I



wherein

X represents a saturated or unsaturated hydrocarbon group containing from 8 to 500 carbon atoms;

Y represents a succinic anhydride residue or an aromatic anhydride residue carried terminally on the group X;

each Z independently represents an organic group carrying an oxygen, sulphur or nitrogen atom through which it is linked to Y;

m is 1 or 2; and

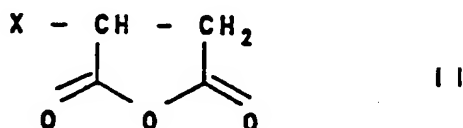
- b) a hydroxyoxime.

The group X is preferably a saturated or unsaturated aliphatic hydrocarbon chain which has a molecular weight of from about 100 to about 8000 and especially from about 100 to about 2000. Thus, the group X preferably contains at least 8 carbon atoms and up to about 150 carbon atoms and can be linear or branched.

Examples of such groups are nonyl, decyl, decenyl, dodecyl, dodecenyl, hexadecyl, octadecyl, octadecenyl and long alkyl chains such as those obtained for example by the polymerisation or copolymerisation of mono-olefins containing from 1 to 6 carbon atoms, for example, ethylene, propylene, but-1-ene, but-2-ene or isobutylene.

Preferred groups X are those derived from the polymerisation of isobutylene. These polymers can be made by standard methods and are commonly referred to as alk(en)yl polymers.

Such polymers have a terminal double bond that can be reacted with maleic anhydride to form alk(en)yl substituted succinic anhydride derivatives by reaction in the presence of a standard condensation catalyst for example a halogen such as bromine, to form a compound of formula II

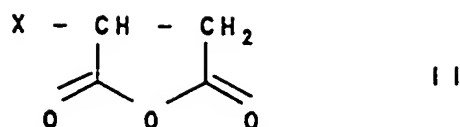


and such compounds are those which are used in the compositions of EP 247728.

The alk(en)yl substituted succinic anhydrides of formula II are commercially readily available and can be used in the form in which they are available without further purification. Polyisobutylene succinic anhydride is commonly referred to as PIBSA.

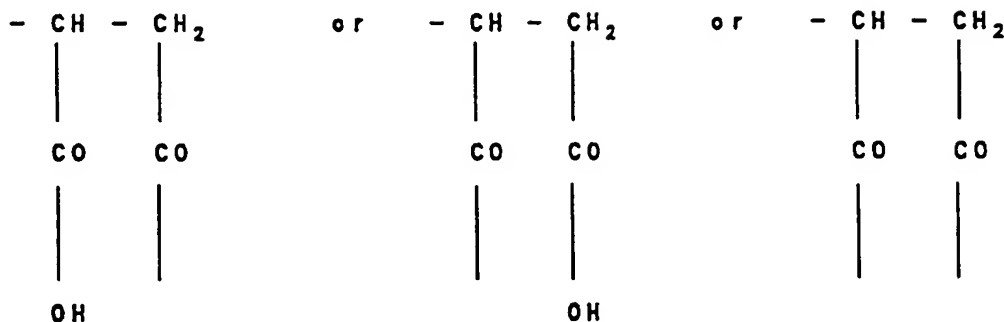
A preferred compound of formula II, is a poly(isobutenyl)succinic anhydride and particularly one with a number average molecular weight of up to 5000.

When the group Y is an aromatic anhydride residue, it is preferably derived from phthalic anhydride, and especially phthalic anhydride wherein the group X is attached in the 4-position relative to the anhydride group. It is preferred, however, that the group Y is a succinic anhydride residue derivable from the succinic anhydride group.



shown in formula II.

When Y is such a group it is preferably a di- or trivalent group of the formula



linking the group X to one or two groups Z.

The group Z in the compound of formula (I) is preferably the residue of an organic compound containing at least one amino or hydroxyl or thiol group and containing up to 20 carbons, more preferably up to 12 carbon atoms and especially up to 6 carbon atoms. The group Z is preferably an optionally substituted alkyl group, which may be linear or branched, but is preferably linear and preferably carries up to a total of 6 amino and/or hydroxyl groups, more preferably from 2 to 4 amino and/or hydroxyl groups, and especially 2 or 3 amino and/or hydroxyl groups. Where the group Z is linked to Y through nitrogen amino this is preferably the nitrogen atom of an amino group which may be primary or secondary but is preferably a primary amino group. It is especially preferred that the organic group contains both an amino group and a hydroxyl group through either of which it may be joined to Y.

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The group Z may contain other groups in addition to the amino and/or hydroxyl and/or thiol groups such as carboxylic acid groups.

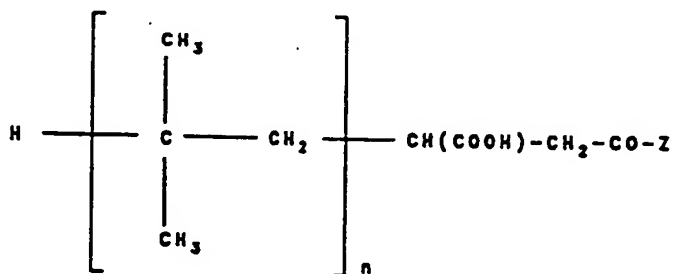
The group Z is preferably derived from an alcohol or a polyol, for example it may be derived from ethylene glycol, propylene glycol, glycerol, trimethylolpropane, pentaerythritol or sorbitol.

The group Z may be derived from an amine or polyamine for example ethylenediamine, trimethylenediamine, hexamethylenediamine, tetraethylenepentamine, dimethylamino-propylamine or diethylamino-propylamine.

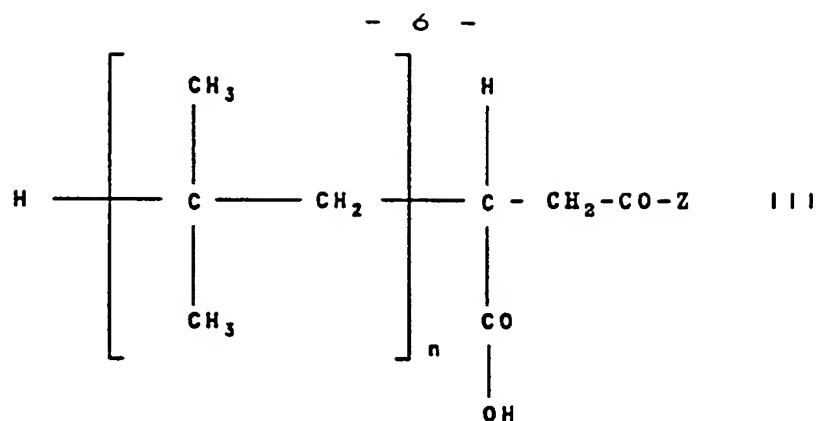
However, the group Z is preferably derived from an organic compound containing both amino and hydroxyl groups such as monoethanolamine, diethanolamine, dipropanolamine, tris(hydroxymethyl)aminomethane or dimethylaminoethanol. The group Z may also be derived from an organic compound containing other groups such as a carboxylic acid group in addition to the amino and/or hydroxyl group, for example glycine or glycollic acid.

In practice the compounds of formula (I) are obtained as a mixture of isomers.

For example, those derived from polyisobutylene and maleic anhydride have the following major components:—



or



wherein

n has an approximate value of from 2 to 100 such that the compound has a molecular weight from about 200 to 5000 and Z is as hereinbefore defined and is linked to the CO group through an oxygen nitrogen or sulphur atom.

The compounds of formula (I) are readily prepared by reacting the appropriate alk(en)yl succinic anhydride with the appropriate organic amino or alcohol compound at a temperature of from 70 to 150°C in an inert solvent or diluent, for example, xylene. Reaction is continued until a substantially constant acid value is achieved.

Preferably, the reagents are used in substantially equimolar proportions based on the anhydride ring and the hydroxyl and/or amino groups.

Thus they produce a half derivative of the succinic group having a free carboxylic acid group obtained by the opening of the anhydride ring, as shown in formulae III and IV.

Excess amounts of the organic moiety represented by Z may be used to react with the alk(en)yl anhydride such that the surface active anhydride derivative of general formula I contains no free carboxylic acid groups.

However, it is preferred that the compound of general formula I contains one free carboxylic acid group, and it is especially preferred that n is 1.

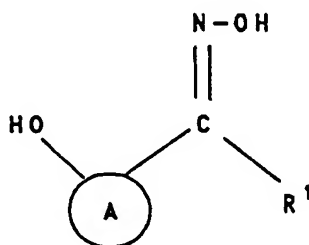


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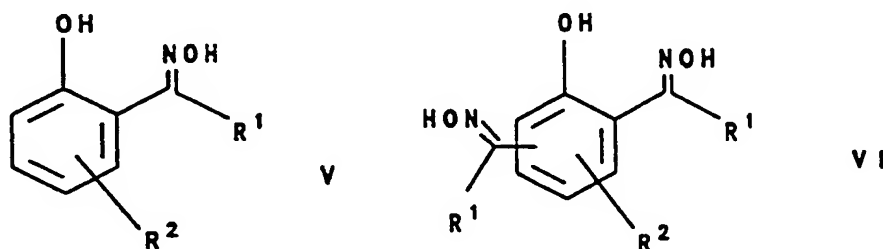
The preferred anhydrides are succinic anhydrides where the group X is an isobutylene polymer of molecular weight from about 100 to 8000, and especially from 600 to 6000 and more especially from 800 to 1200.

A preferred example of the compounds of general formula I is the reaction product of polyisobutenyl succinic anhydride having a molecular weight of approximately 1100 and ethanolamine.

The hydroxyoxime which is component (b) of the composition in accordance with the present invention is preferably an aromatic hydroxyoxime of the formula



wherein A is an aromatic system, especially a benzene ring; and R<sup>1</sup> is a hydrogen atom or a substituted or unsubstituted hydrocarbon group. The hydroxyoxime is more preferably a hydroxy monooxime of the formula V or hydroxy bisoxime of formula



wherein R<sup>1</sup> is as hereinbefore defined; and R<sup>2</sup> is a substituted or unsubstituted C<sub>5</sub>-22 hydrocarbon group.

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The group R<sup>1</sup> is preferably hydrogen but may be an alkyl, aryl or benzyl group, which may be substituted. When R<sup>1</sup> is aryl, it is preferably phenyl, and when R<sup>1</sup> is alkyl, it is preferably C<sub>1-4</sub> alkyl, for example methyl.

5 In the bis-oxime compound of formula VI, the second oxime group is preferably in the 6-position (ie ortho to the hydroxyl group) and especially preferred compounds of this type are the 2,6-bis(oximinomethyl)-4-alkylphenols.

10 The group R<sup>2</sup> is preferably a C<sub>5-14</sub> alkyl group which may be linear or branched and may be saturated or unsaturated, but is preferably saturated. The group R<sup>2</sup> is preferably in the para position with respect to the phenolic hydroxyl group when the compound is a 2-hydroxy-5-alkylbenzaldoxime or 2-hydroxy-5-alkylbenzoketoxime. Examples of the group R<sup>2</sup> include decyl, decenyl, dodecyl, dodecenyl, hexadecyl, 15 octadecenyl and especially nonyl. Specific examples of the hydroxyoxime compounds are 2-hydroxy-5-nonylbenzaldoxime, 2-hydroxy-5-dodecylbenzaldoxime and 2-hydroxy-5-nonylaceto-phenoneoxime.

20 The weight ratio of the surface active anhydride derivative of general formula I to the hydroxyoxime may be between 99:1 to 1:99 but will generally be from 10:1 to 1:10, particularly from 5:1 to 1:5. However, the preferred proportions will be dependent on the particular compounds present in the mixture.

25 The compositions of the present invention provide improved corrosion inhibition in corrodible metals and certain of the compositions also provide improved anti-wear properties and/or improved anti-oxidation properties.

30 Thus, as a further aspect of the present invention, there is provided a process which comprises treating at least part of the surface of a metal with a composition comprising (a) a surface active anhydride derivative of general formula I, and

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(b) a hydroxyoxime,  
as hereinbefore defined.

The process of the present invention is especially  
suitable for the corrosion inhibition of iron, zinc, copper,  
tin and aluminium and in particular mild steel and the zinc  
surface of galvanised steel.

The metal may be treated directly with the composition.

However it is generally preferred to apply the  
composition to the metal surface in the form of a solution in  
a suitable organic liquid or as an aqueous emulsion or  
dispersion of the composition or as an aqueous emulsion of a  
solution of the composition in an organic liquid.

The composition may be used to provide a protective  
coating in its own right, or as a metal pre-treatment before  
the application of a surface coating, or may be incorporated  
into a surface coating composition, or may be employed as an  
additive to refined petroleum products such as lubricating  
oils, turbine oils, fuel oils, gasohols and greases.

Any suitable organic liquid may be used as a solvent for  
the composition, such as aliphatic and aromatic hydrocarbons,  
and their halogenated derivatives, ethers and ketones.  
Especially preferred solvents are those having good wetting  
and drying properties and include for example benzene,  
toluene, xylene, kerosene, white spirit, chloroform, and  
1,1,1-trichloroethane.

Aqueous emulsions of the composition may be formed in  
conventional manner using conventional dispersants and  
surfactants, including non-ionic dispersants.

It may be convenient to treat the metal with an aqueous  
emulsion of the composition, particularly if the anhydride  
present in the composition is a liquid which can be formed  
directly into an emulsion.

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5 The process of the present invention may provide corrosion inhibition either without the application of a further surface coating or as a pre-treatment before the application of a further surface coating. Thus, the treatment may be used for example as a temporary protection of a metal, for example while the metal is being transferred from one site to another and the protective coating subsequently removed before or during further processing.

10 Alternatively, the composition may be formulated in a surface coating composition, for example a paint (primer) such as air-drying, oil-modified system or a system including a chlorinated rubber, lacquer, resin or other protective coating.

15 The surface coating may be a solvent-based composition, for example a cellulose/solvent based primer paint such as those used for car "touch up" paints.

20 The components of the composition are soluble in solvents generally used for such primers (for example nitrocellulose) and may be incorporated directly. The composition may also be used as an emulsion in aqueous emulsion surface coating systems, for example primers or protective coatings based on polymer latices such as for example acrylic and styrene/acrylic latices and vinyl acrylic co-polymer latices including acrylate modified vinyl chloride vinylidene chloride copolymer latices.

25 The composition may also for example be incorporated in temporary alkali-removable protective coatings (whether solvent-based or emulsion based) of the addition polymer type in which the polymer contains carboxyl groups.

30 The composition or the solution or emulsion thereof may be applied to the metal in conventional manner, for example by dipping, spraying or brushing. The temperature of the application may be from 0 to 50°C.

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Typically, solutions of the composition may contain from 0.1 to 20% by weight of the mixture of the anhydride derivative and the hydroxyoxime, whilst emulsions preferably contain from 0.1 to 5% by weight of the mixture of anhydride derivative and hydroxyoxime.

The presence of from 0.1 to 2% by weight of the mixture of anhydride derivative and hydroxyoxime in a surface coating emulsion formulation is generally sufficient to provide improved corrosion inhibition.

If the composition contains materials in addition to the anhydride derivative and the hydroxyoxime, the amount of the composition which is used should be sufficient to provide the mixture of the anhydride derivative and the hydroxyoxime in the proportions as set out herein.

The metal to be treated may be brightly polished and/or freshly cleaned, but it is an advantageous feature of the process of the present invention that effective corrosion inhibition may be obtained even on a lightly rusted surface. Indeed we have found that better results are in general achieved when a surface is treated in an "as received" condition than when that same surface is freshly cleaned or brightly polished.

The present composition has been found to give greatly improved protection of metal surface against wear and corrosion in tank oil systems and greases which are subject to contact with water.

Thus, according to a further aspect of the present invention there is provided a lubricant composition comprising

a) a surface active anhydride derivative of the formula 1



wherein X, Y, Z and m are as hereinbefore defined;

b) a hydroxyoxime; and

c) an oil or a grease;

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The oil is preferably a mineral oil or a synthetic oil such as a polyalkylene glycol, a polyalpha olefin, an ester and in particular a phthalate, a perfluoroalkyl ether or a silicone.

5 The greases are preferably mineral or synthetic oils which have been thickened by the addition of gelling agents.

Typically, the mineral oil is a hydrocarbon oil, and the synthetic oil may be for example a synthetic hydrocarbon oil, a diester such as di(2-ethylhexyl)sebacate, a perfluoroalkyl  
10 ether or a silicone oil.

Many greases are obtained using soaps as the gelling agent, especially lithium soaps. However, non-soap gelling agents can produce a grease having improved properties, especially the clarity to remain effective at higher use  
15 temperatures. Examples of non-soap gelling agents present in such greases are clays, carbon black, silica and polyurea all of which are preferably incorporated into the oil as finely divided solid materials.

Finely divided clay particles, for example clays of the bentonite or hectorite types can be used as non-soap gelling  
20 agents to obtain a grease from a mineral oil. Preferably, the clay particles are surface-coated with an organic material such as a quaternary ammonium compound.

Where the grease is based on a silicone oil, the non-soap gelling agent is preferably silica, and especially  
25 fused silica having an average particle size less than one micron.

The lubricant composition preferably contains from 0.1 to 30% by weight of the hydroxyoxime relative to the total  
30 volume thereof and more preferably from 0.1 to 5%.

In addition to the hydroxyoxime and anhydride derivative, the lubricant composition may include various other ingredients commonly incorporated into such compositions such as oxidation inhibitors, and extreme pressure additives.

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The lubricant composition may be prepared using any of the techniques which are effective for incorporating additives into oil or grease media.

For example a grease composition may be formed by stirring together the thickened grease with a composition containing the anhydride derivative and the hydroxyoxime.

As an alternative, the composition of the anhydride derivative and the hydroxyoxime may be incorporated into an oil which is subsequently thickened to form a grease.

The lubricant composition in which component (c) is a grease may be used in any application for which a grease is known, and in particular can be used in general automotive applications and also in bearings including high performance bearings.

We have found that when subjected to an anti-rust test the present lubricant composition shown improved resistance to corrosion and destructuring compared to a grease containing the composition disclosed in EP 247728.

A bearing containing, a lubricant composition in accordance with the present invention is a further feature of the present invention.

The compositions of the invention may also include other materials, particularly materials which also provide corrosion inhibiting properties. Such corrosion inhibiting materials include, for example, petroleum sulphonates, aryl sulphonates and the metal salts thereof.

The other materials may be present in the composition in a substantial proportion of up to several times, for example up to ten times, by weight of the composition of the anhydride and the hydroxyoxime.

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In general no particular advantage is achieved by using a large excess of the other materials and preferably the proportion of the other materials do not exceed twice the weight of the composition and conveniently the other materials are present in essentially an equal proportion by weight relative to the weight of the composition.

Various aspects of the present invention are set out in more detail hereafter in the following illustrative examples in which all parts and percentages are by weight unless otherwise stated.

#### Example 1

Samples of a solvent-neutral mineral oil, containing 0.1% corrosion inhibitor and a sample of the sample oil containing no additive were subjected to an oil/water corrosion test based on ASTM D 665B modified as set out hereafter.

The test involves stirring a mixture of the oil (300 ml.) and synthetic sea water (30 ml.) for 24 hours at 1000 rpm and at a temperature of 60°C containing a completely immersed and previously weighed cylindrical steel specimen.

The metal specimen is then removed, cleaned, re-weighed to determine weight loss (if any) and examined for evidence of corrosion.

The results obtained are set out in Table One.



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TABLE ONE

Example or Comp Ex	Additive (a)	Wt. loss (mg)	Visual Appearance
1	10.05% BAO + 0.05% PMEAI	0.2	Rust free
A	0.1% BAO	113	Corroded
B	0.1% PMEAI	60	Corroded
C	Nil	106	Corroded

## Notes to Table One

(a) BAO is 2-hydroxy-5-nonylbenzaloxime

PMEA is a reaction product of polyisobutenylsuccinic anhydride of approximate molecular weight 1100 and monoethanolamine.

## Example 2

Samples of a lithium complex grease containing 2% corrosion inhibitor and also a sample of the same grease containing no additive were subjected to a modified ASTM D1831 (Roll Stability of lubricating grease) test as described hereinafter.

A sample of lithium complex grease (90 parts) containing 2% corrosion inhibitor and distilled water (10 parts) were placed in a test cylinder.

The grease was distributed evenly over the inside wall of the cylinder, a weighted roll was placed in the cylinder and the cap tightened. The cylinder was then rolled at 165 ± 15 rpm for two hours after which the grease removed and subjected to the cone penetration test for lubricating greases.

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This was carried out using one-half scale cone equipment and the procedure of ASTM D1403. The cone penetrations were carried out on samples of grease which had been brought to 25°C and subjected to sixty double strokes in a grease worker in the manner described in ASTM D1403. Using the standard formula as set out in ASTM D1403, the measurements made were transformed to give the work penetration of the cone for full scale cone equipment.

The results obtained are set out in Table Two.

TABLE TWO

Example or Comp. Ex	Additive (a)	Worked Penetration (b)
2	1.5% BAO + 0.5% PMEA	311
D	1.5% BAO + 0.5% TPSA	destructured
E	Nil	300

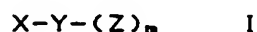
## Notes to Table Two

- (a) BAO is 2-hydroxy-5-nonylbenzaloxime  
PMEA is a reaction product of polyisobutenylsuccinic anhydride of approximate molecular weight 1100 and monoethanolamine  
TPSA is tetrapropenylsuccinic anhydride.
- (b) measured in tenths of a millimetre.

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## CLAIMS

1. A composition comprising  
a) a surface-active anhydride derivative of the general  
formula I



wherein

X represents a saturated or unsaturated hydrocarbon chain containing from 8 to 500 carbon atoms;

Y represents a succinic anhydride residue or an aromatic anhydride residue carried terminally on the group X;

each Z independently represents an organic group carrying an oxygen, sulphur or nitrogen atom through which it is linked to Y; and

m is 1 or 2, and

- b) a hydroxyoxime

2. A composition as claimed in claim 1 wherein m is 1, and Y represents a succinic anhydride residue.

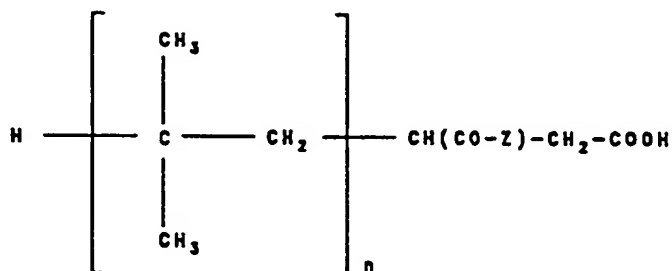
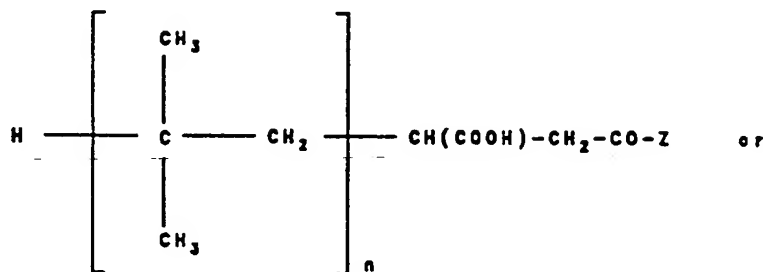
3. A composition as claimed in claim 1 wherein the anhydride derivative is one in which X is nonyl, decyl, decenyl, dodecyl, dodecenyl, hexadecyl, octadecyl or octadecenyl, or in which X is derived from an isobutylene polymer made from but-1-ene, but-2-ene or isobutene and having a molecular weight from about 800 to 2000.

4. A composition as claimed in claim 1 wherein the group : is derived from ethylene glycol, propylene glycol, glycerol, trimethylolpropane, pentaerythritol, sorbitol, ethylenediamine, trimethylenediamine, hexamethylenediamine, tetraethylenepentamine, dimethylaminopropylamine, diethylaminopropylamine, ethanolamine, diethanolamine,

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dipropanolamine, tris(hydroxymethyl)aminoethane or dimethylaminoethanol.

5. A composition as claimed in claim 1 wherein the anhydride derivative is a complex mixture of isomers having as major components



wherein

n has an approximate value of from 2 to 100 such that the compound has a molecular weight from about 200 to 5000; and Z is a straight or branched alkyl group carrying hydroxyl, amino and/or carboxylic acid substituents, the alkyl group being bonded to the CO group through the residue of a hydroxyl or amino group.

6. A composition as claimed in claim 1 which contains a hydroxyoxime of general formula

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wherein

A is an aromatic system; and

R' is a hydrogen atom or a substituted or unsubstituted hydrocarbon group.

7. A composition as claimed in claim 6 wherein the hydroxyoxime is 2-hydroxy-5-nonylbenzaldoxime or 2-hydroxy-5-nonylacetophenone oxime.

8. A process which comprises treating at least part of the surface of a metal with a composition as claimed in any one of claims 1 to 7.

9. A metal at least part of one surface of which has been coated with a composition as claimed in any one of claims 1 to 7.

10. A lubricant composition comprising

a) a surface-active anhydride derivative of the general formula I



wherein

X represents a saturated or unsaturated hydrocarbon chain containing from 8 to 500 carbon atoms;

Y represents a succinic anhydride residue or an aromatic anhydride residue carried terminally on the group X;

each Z independently represents an organic group carrying an oxygen, sulphur or nitrogen atom through which it is linked to Y; and

m is 1 or 2.

b) a hydroxyoxime; and

c) an oil or a grease

## INTERNATIONAL SEARCH REPORT

Inter. Application No

PCT/GB 93/01473

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 5 C10M141/06 C23F11/10

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 5 C10M C23F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP,A,0 247 728 (IMPERIAL CHEMICAL INDUSTRIES) 2 December 1987 cited in the application see page 1, line 42 - page 5, line 13; claims 1-5,7-14 ---	1-3,6-10
A	EP,A,0 208 560 (EXXON CHEMICAL PATENTS INC.) 14 January 1987 see page 4, line 1 - page 13, line 34; claims 1,4-11,17-22; example 11 ---	1-4,8-10
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☐ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

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2 November 1993

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

Inter. Application No  
PCT/GB 93/01473

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